A theoretical analysis of the thickness dependence of the localization effect on the normal-state resistivities in high-T_c $Y_1Ba_2Cu_3O_{7-\delta}$ thin films

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(Received 11 October 1991; accepted for publication 24 February 1992)

We have demonstrated that the normal-state resistivity of sufficiently thick $Y_1Ba_2Cu_3O_{7-\delta}$ thin films can be fitted very well by the Bloch–Gruneisen equation. As the film thickness decreases, the localization effect becomes predominant presuming the manifestation of surface defects and vacancies over the bulk region. The evaluated Debye temperatures span over a range which is consistent with experimental values. Below some critical thicknesses, the transport behavior in thin $Y_1Ba_2Cu_3O_{7-\delta}$ films deviates from the Boltzmann conductivity and the temperature coefficient of the electric resistivity becomes negative, which can be explained by adopting quantum corrections for localization and interaction effects. The origin of nonmetallic behavior can be understood quantitatively in terms of the competition between the quantum corrections and the conventional Boltzmann conductivity. In these ultrathin superconducting films, the elastic mean-free path arising from defect and heavy impurity scattering is very short and much smaller than the inelastic mean-free path, even at room temperature.

I. INTRODUCTION

The variation of normal-state resistivity of high-T_c oxide superconductors is linear within a wide temperature region. As reported in the paper of Meyer *et al.*,[1] the temperature coefficient of the electrical resistivity of $Y_1Ba_2Cu_3O_{7-\delta}$ thin films deposited by inverted cylindrical magnetron sputtering onto (100) $SrTiO_3$ substrates changes from a positive value to negative as the thickness of films decreases below 30 Å. The superconductivity even disappears when the film thickness is below some critical value. As shown in Fig. 1 of Ref. 1, the normal state resistivities for films of 1000, 50, and 33 Å are metallic and become nometallic when the thickness is reduced below 30 Å. Since the accuracy of the average thickness determination is estimated to be about 10%, the abrupt changes of the transport properties below a critical film thickness do not indicate an intrinsic characteristic of the material caused by the film thickness partly because the mean-free path of the charge carriers in $Y_1Ba_2Cu_3O_{7-\delta}$ is very short. The transport in the degraded material is temperature activated, leading to nometallic behavior superimposed on a partial superconducting transition at $T<T_c$. The structures of the ultrathin films may show a mixture of superconducting grains surrounded with defects and oxygen vacancies. The supercurrents can percolate through the normal boundaries, though the normal resistivities exhibit disordered behavior.

II. NORMAL STATE RESISTIVITY

For good metals, the elastic scattering time is predominantly determined by phonon scattering, not by impurity or defect scattering, and the resistivity follows the Bloch–Gruneisen law. In this case, elastic and inelastic scatterings are contributed from acoustic and optical phonons, respectively. Formulating the interaction matrix in terms of phonon creation and annihilation processes and allowing for energy lost or gained by carriers, the Bloch–Gruneisen equation is described as[2]

$$\rho(T)=\rho_0+4\rho'\frac{T^5}{\theta^4}\int_0^{\theta/T}\frac{z^5\,dz}{(e^z-1)(1-e^{-z})},\qquad(1)$$

where ρ_0 is the residual resistivity, θ is the Debye temperature, and ρ' is a constant specifying the high-temperature limit. At high temperatures, where θ/T is small, the integral approaches to $(\theta/T)^4$, and $\rho(T)\propto T$.

The normal-state resistivity of $Y_1Ba_2Cu_3O_{7-\delta}$ thin films, in which the temperature coefficient is positive, can be fitted very well with Eq. (1) by using the method of the least mean squares. In the fitting process, the ρ_0 and ρ' are treated as adjustable parameters and the Debye temperature θ is kept constant. Figure 1 shows the relation between the Debye temperature and the sum of squares of errors (SSE). The best fitting corresponds to a value of θ within the range 200–250 K for the film thickness of 1000 Å and within 300–330 K in the cases of 50 and 33 Å. The choice of various values of θ only causes the correct value of ρ_0 to fluctuate. The experimental data for the Debye temperature for the $Y_1Ba_2Cu_3O_{7-\delta}$ superconductor is temperature dependent and its value is between 460 and 520 K for the temperatures between 100 and 300 K.[3] The dependences of θ and ρ' and SSE are weak. As shown in Fig. 1, the slight increase of SSE corresponding to the use of the experimental θ rather than that from the best fitting is acceptable. Table I lists the results of best fitting for ρ_0 and ρ' at different film thicknesses. Irrespective of the violent change of the residual resistivity, the other specifying constants increase slowly as the film thickness decreases. The larger value of ρ_0 is a synonym of the shorter value of the mean-free path (l_0) reflecting the manifestation of defect or impurity scattering as the film thickness decreases.

Exploiting the best fitting parameters, the theoretical Bloch–Gruneisen expression can congruently match the experimental data, as shown in Fig. 2. Inspection of Table

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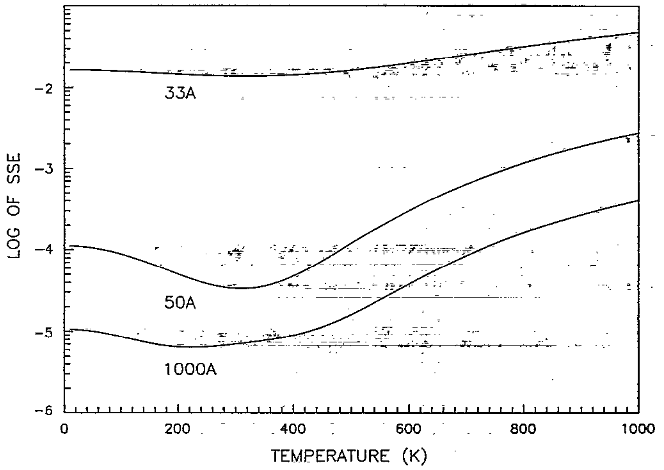

FIG. 1. The sums of squares of errors for thick superconducting thin films with a different Debye temperature. There are ten experimental data points used for the fitting.

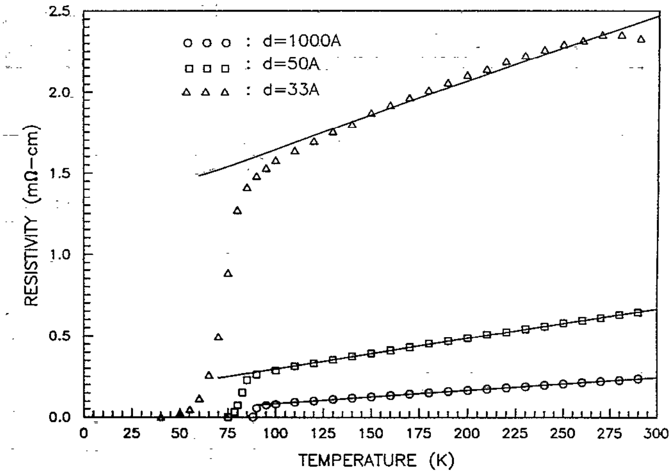

FIG. 2. The dependences of electrical resistivity on temperature with film thicknesses being 1000, 50, and 33 Å. The solid lines are the result of the best fitting with the Bloch–Gruneisen equation. The symbols are the experimental data extracted from Fig. 1 of Ref. 1.

I and Fig. 2 show that the sum of squares of errors becomes larger as the film thickness becomes thinner.

III. WEAK LOCALIZATION

When the film thickness gets thinner than 30 Å, the normal-state resistivity reveals a nonmetallic behavior. This implies that there may exist localized states in the ultrathin films. Localization causes the carrier wavefunction to fall off as a power law. The conductivity which deviates from the Boltzmann conductivity (σ_b) can be derived from the Kubo–Greenwood formulation, which yields[4]

$$\sigma \simeq \left| \int_0^{E_F} \Psi_E \left(\frac{\partial}{\partial X}\right) \Psi_{E'}\, dX^3 \right|^2$$

where the integral takes the average over E, E' below energy E_F. Following Kaveh and Mott,[4] the quantum correction due to the power law localization for disordered noninteracting carriers is given by[5,6]

TABLE I. (a) The best fitting results of ρ_0, ρ', SSE, and θ for different film thicknesses of 1000, 50, and 33 Å. (b) The fitting results for the Debye temperature within 460–520 K which are the experimental values at temperatures between 100 and 300 K.

d (Å)	ρ_0(mΩ cm)	ρ'(10^{-7} Ω cm)	SSE(10^{-4})	θ (K)
(a)				
1000	0.027–0.034	0.72–0.74	0.0642	200–250
50	0.192–0.202	1.64–1.67	0.335	290–330
33	1.405–1.425	3.69–3.75	138	300–340
(b)				
1000	0.064–0.071	0.68	0.12–0.19	460–520
50	0.192–0.202	1.59	0.80–1.44	460–520
33	1.405–1.425	3.58	152–168	460–520

$$\delta\sigma_l(T) = \frac{e^2}{\pi^2\hbar}\left(\frac{1}{L_{\text{in}}} - \frac{1}{l_0}\right), \tag{2}$$

where the diffusion length due to inelastic scattering at finite temperature is defined as

$$L_{\text{in}} = \sqrt{\frac{l_0 l_{\text{in}}}{2}}.$$

The localization effect exists only when $l_0 < L_{\text{in}}$, i.e., $\delta\sigma_l(T) < 0$. If $l_{\text{in}} < l_0$, then $\delta\sigma_l = 0$.

By including the correlation effect, the carrier interaction contributes a conductivity similar to Eq. (2) and is expressed as[5,7]

$$\delta\sigma_{\text{int}}(T) = \frac{e^2}{\pi^2\hbar} f(x) \left(\frac{1}{L_{\text{int}}} - \frac{1}{l_0}\right), \tag{3}$$

where the interaction length is defined as

$$L_{\text{int}} = \sqrt{\frac{\hbar D}{k_B T}}$$

and

$$f(x) = 1 - \frac{3}{2x}\ln(1+x), \quad x = (2k_F \cdot \lambda)^2$$

The k_F, λ, and D denote the Fermi wave vector, the Thomas screening length, and diffusion constant, respectively. Equation (3) is constrained by the same conditions as Eq. (2) with $L_{\text{int}} > l_0$, but its sign is decided by $f(x)$. The function $f(x)$ is the combined result of exchange and Hartree interaction terms.[8] If λ is much larger than the interparticle spacing, then only the exchange term need be counted and $f(x) \to 0$. However, as $k_F\lambda$ becomes small, the Hartree term begins to intervene and the sign of $f(x)$ may be changed from positive to negative.

Including all the quantum corrections, the total conductivity becomes

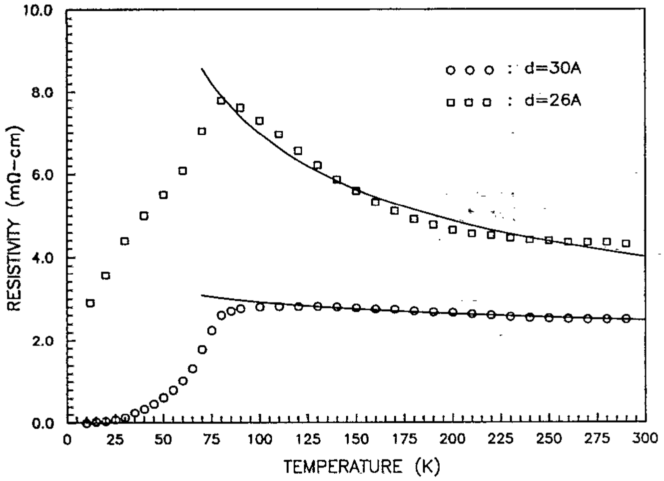

FIG. 3. The dependences of electrical resistivity on temperature for film thicknesses of 30 and 26 Å. The solid lines are obtained by using Eq. (4) with the best fitting parameters. The symbols are also the experimental data extracted from Fig. 1 of Ref. 1.

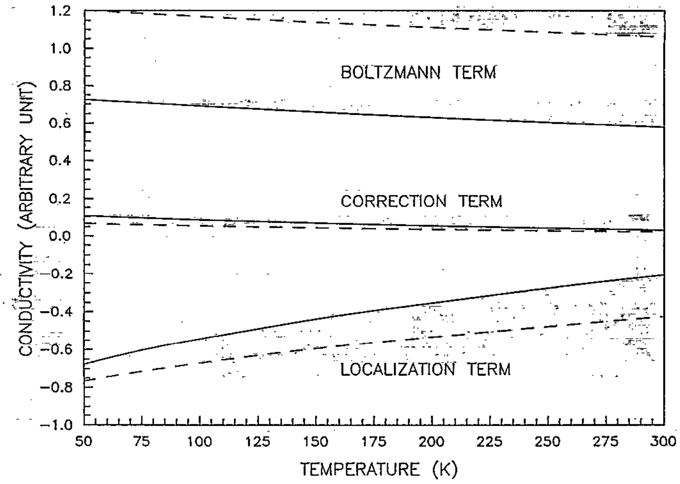

FIG. 4. The calculated different components of total conductivities for film thicknesses are 30 and 26 Å, based on Eqs. (2)–(4). The Bloch–Gruneisen solid lines are the classical Boltzmann conductivities. The dash and dot lines denote the localization and interaction terms of quantum correction, respectively.

$$\begin{aligned}\rho^{-1}=\sigma&=\sigma_b+\delta\sigma_l+\delta\sigma_{\rm int}\\&=\frac{e^2}{\pi^2\hbar}\frac{1}{l_0}\left[\frac{(k_Fl_0)^2}{\cdot 3}\left(1+\frac{l_0}{l_{\rm in}}\right)^{-1}+\left(\sqrt{\frac{2l_0}{L_{\rm in}}}-1\right)\right.\\&\quad\left.+f(x)\left(\sqrt{\frac{k_BT}{\hbar D}}\cdot l_0-1\right)\right].\end{aligned}\tag{4}$$

IV. COMPARISON TO EXPERIMENT

The balance between these competitive effects on the current transport determines the sign and the size of $d\rho/dT$. In Eq. (4), the first term in the brackets is the Boltzmann conductivity resulting from conventional thermal excitation of various scattering processes with free carriers. Usually, the temperature dependence of $l_{\rm in}$ is expressed by $l_{\rm in}(T)=A\cdot T^{-p}$, where A is a constant and the value of p can vary with temperature (e.g., $p=2-5$ for $T<40$ K, $p=1$ for $T>200$ K). Since we are interested in the temperature region of 100–300 K, the ratio of $l_0/l_{\rm in}$ is proportional to T, due to the optical-phonon scattering mechanism. The theoretical fitting obtained by implementing Eq. (4) for film thicknesses of 30 and 26 Å are shown in Fig. 3. The quantum corrections become important as the film thickness decreases implicit in the condition $k_F\cdot l_0\sim 1$. We have decomposed the quantum correction into its component terms in Eq. (4), as shown in Fig. 4, which shows that the localization term of the quantum correction is negative, whereas the interaction term is positive. The lower the temperature, the larger the quantum correction. However, the temperature dependence of conductivity is dominantly controlled by the localization effect in the ultrathin films, which makes them behave as nonmetallic. The best fitting parameters are tabulated in Table II.

From Table II, we find that the elastic mean-free path l_0 and diffusion coefficient D almost remain constant. The value of l_0 is very short, of the order of interatomic spacing. Even at room temperature, l_0 is much smaller than $l_{\rm in}$. The smallness of l_0 justifies the quantum corrections. On the other hand, the Fermi wave vector k_F, and inelastic mean-free path $l_{\rm in}$, diminish as the film thickness is reduced. It is apparent that k_Fl_0 decreases and the power law component becomes important as the disorder increases, which leads to the localization quantum correction to the classical Boltzmann conductivity.

V. CONCLUSIONS

In conclusion, the normal-state resistivities of thick $Y_1Ba_2Cu_3O_{7-\delta}$ thin films follow the Bloch–Gruneisen equation. The residual resistivity increases as the film thickness decreases, suggesting that the films become disordered as they get thinner. For the ultrathin films, with thickness below 30 Å, the resistivity contributed by the degraded surface will overwhelm that due to the bulk region underneath which is assumed to act in parallel in the conduction, and the localization and interaction effects begin to intervene. If the temperature dependence of resistivity is governed by the localization effect, the temperature coefficient of electric resistivity is negative. The localization effect reduces the Boltzmann conductivity while the interaction effect may enhance the conductivity depending on the value of the carrier density. For resistivity near critical temperature, the excess conductivity due to superconducting pairs may play a role and can be described by the Aslamazov and Larkin equation.[9,10]

TABLE II. The best fitting results of l_0, $l_{\rm in}$, k_F, and D for the thicknesses of 30 and 26 Å.

d(Å)	30	26
k_F(1/Å)	0.480–0.474	0.377–0.365
l_0(Å)	4	4
$l_{\rm in}$(Å)	$7307/T$–$5255/T$	$3817/T$–$2883/T$
D(cm²/s)	0.1050–0.0556	0.1007–0.05

ACKNOWLEDGMENT

This work was supported by the National Science Council of the Republic of China under Contract NSC 80-0212-M-007.

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